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16. Abstract

During this program MetroLaser pioneered the development of Resonant Holographic Interferometry (RHI); a holographic technique capable of recording the concentration of a target species in complex flame environments. Some of the attributes that make RHI attractive to the combustion community are:

- Operates on ground state transitions (insensitive to quenching).
- Simultaneous imaging of gaseous species and solid or liquid object.
- Background free signals.

The program targeted the concentration measurement of species with resonant transitions in the visible (Na), ultraviolet (OH), and near infrared (K) spectral regions. This broad operating range required the development of auxiliary technologies such as real-time holographic recording and reconstruction using nonlinear optical materials.

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RESONANT HOLOGRAPHIC INTERFEROMETRY, AN INNOVATIVE TECHNIQUE FOR COMBUSTION DIAGNOSTICS

FINAL REPORT

Peter A. DeBarber, Ph.D.
and
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December 31, 1995

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PROBLEM STUDIED

The Army Research Office has a goal of supporting projects leading to an increased fundamental understanding of combustion and propulsion phenomena. Most military and commercial endeavors rely heavily upon energy conversion for the generation of power. The mainstay of these energy conversion processes is combustion. Efforts to gain a more detailed understanding of the physics and chemistry of combustion are needed to increase the efficiency of these processes. These efforts take on the form of experiment and theory. Modeling is a more cost effective approach at improving combustor design and technology than empirical design; however experiments are still very much needed to validate the theoretical models and to study complex combustors which cannot be analyzed with current models. Particularly useful parameters to the modelers and developers are temperature, species concentration, bulk flowfield density, and velocity. In addition, highly resolved spatial profiles of these parameters are sought for model validation. To this end, MetroLaser has developed a new class of advanced optical diagnostics.

During the past three years, MetroLaser embarked on theoretical and experimental efforts to develop new optical diagnostics applied to combustion research. Our particular program with the ARO focused on the development of an entirely new diagnostic technology called Resonant Holographic Interferometry (RHI). Since most chemical species of importance in combustion exhibit absorptions in the visible, ultraviolet (UV), or near infrared (IR) wavelength regions, our development efforts explored RHI applications in each of these areas. Our three year program with the ARO progressed in the following way. In the first year, we examined visible RHI applications. In the second year, we expanded our measurements into the UV wavelength regime. And in the third year, we researched near IR applications. This Final Report summarizes our work in each of these spectral areas. The report begins with a brief treatment on the theoretical background and conceptual design of RHI. We next highlight our results for each of the three wavelength regions, visible, UV, and near IR. We also identify technical opportunities discovered in the course of this research in spray combustion applications, stimulated Brillouin scattering, and holographic data reduction. We conclude this report with an assessment of the state-of-the-art of RHI.

RESONANCE HOLOGRAPHIC INTERFEROMETRY (RHI) THEORETICAL BACKGROUND

RHI is a laser-based diagnostic which combines the three-dimensional imaging capability of holography, the phase sensitive detection of interferometry, and the species specificity of resonance spectroscopy to form a powerful, versatile diagnostic package[1]. RHI provides researchers a method of obtaining nonintrusive measurements in environments that are characterized by a high degree of scattering, luminosity, and/or optical density.. This is accomplished by recording two simultaneous holograms at two different laser wavelengths: one tuned near an absorption line, and the other tuned off this feature. Upon reconstruction, the resulting interference fringes correspond uniquely to the density of the species under interrogation because phase contributions from background species, thermal and pressure instabilities, and optical aberrations are automatically subtracted out.

The ability to distinguish individual species is important for many diagnostics applied to combustion environments. Conventional holographic interferometry measures the total non-resonant phase contributions from all species present which are inseparable from each other as well as the temperature and pressure gradients present. However, with RHI, the observation of minor combustion species in the presence of major background species and large temperature and pressure gradients is possible because of the large variation in the index-of-refraction near an absorption resonance. Thus, by utilizing the resonant refraction with the background subtraction inherent to holographic interferometry, RHI becomes a valuable species-specific combustion diagnostic.

Below we briefly outline the RHI phenomenology and experimental design concept. For a more indepth coverage of the theoretical underpinnings, the reader is directed to publications included in the Bibliography section of this report.

RHI Phenomenology

The RHI technique enables the measurement of the phase change experienced by a coherent laser beam as it propagates due to changes in the real part of the refractive index of the medium. The raw data is in the form of resonant holographic interferograms that are comprised of fringe patterns. The fringes represent contours of constant optical pathlength due to the chemical species under interrogation.. These contours are converted to species number density profiles. The RHI method takes advantage of the dramatic change in the index-of-refraction in the vicinity of a resonant absorption feature. The method requires two laser wavelengths. Thus, by tuning one laser near the resonant feature and the other off resonance, a *species specific* hologram can be generated. The near-resonance laser beam is used to make a hologram recording the phase of the resonant species plus all of the background phase effects as well. The off-resonance laser beam is used to make a hologram recording of only the background phase effects. When the two superimposed holograms are reconstructed, an interferogram results due the phase difference between the two holograms. Since the only difference between the two holograms is the resonant phase shift, the resultant fringe pattern is attributable uniquely to the resonant species under interrogation.

An alternative approach used to increase RHI sensitivity by a factor of two is implemented by tuning each laser to the corresponding maximum and minimum positions in the dispersion curve. Since the background contribution to the phase is constant over this range, and the resonant contribution to phase is additive, the phase difference between the two waves is simply the sum of the absolute value of the phase shift due to the resonance effect. At the maximum and minimum positions, the absolute value of the phase shift is equal, and therefore the phase difference between the two waves is twice the phase shift due to any one wave.

In addition to phase information that results from changes in the real part of the index-of-refraction, RHI also stores absorption information that results from changes in the imaginary part of the index-of-refraction. This ability to store and measure multiple spectroscopic parameters is referred to as "multiplex spectroscopy". Another possible implementation of RHI is to access two

separate transitions near-resonance. This provides information regarding the species' internal state populations which allows the computation of temperature by assuming a Boltzmann distribution.

A master equation predicting RHI fringe shifts can be written in a straightforward way. The pathlength through the target species can and does change across an RHI field-of-view. The phase shift difference experienced by the two object beams is maximized by symmetrical tuning of the two lasers about the absorption feature at the half-maximum intensity positions.

The RHI fringe shift, *F*, is given by the integrated phase difference between the two recorded holograms divided by 2π,

$$F = 2\,K f L N \frac{g^{*}_{dis}\ \omega_o, \omega_l}{2\pi} \quad . \tag{1}$$

In the above, we have used the following symbols:

K	a constant (2.24×10^{-14} cm)
f	species oscillator strength (dimensionless)
L	pathlength through target species (cm)
N	target species number density along path L (cm^{-3}) .

The function $g^{*}_{dis}(\omega_o, \omega_l)$ (units of cm) denotes the weighted, normalized, real part of the refractive index of the target species. The factor of two in the numerator accounts for symmetrical tuning of the RHI probe lasers about the absorption line center.

RHI Experiment Design

The RHI instrument employs two independently tunable narrow-band short pulsed laser sources. The independent tunability gives researchers maximum flexibility in selecting resonance transitions, and in tuning to the on- and off-resonance wavelengths. This also allows access to two separate transitions for the possibility of measuring temperature. The narrow band feature provides two benefits. The first is the increase in instrument sensitivity with decreasing laser linewidth. The second is the concomitant increase in the beam coherence length. Longer coherence lengths facilitate matching of the object and reference beam path lengths. Path matching within one coherence length is a requirement for high quality holograms. The short pulse feature provides excellent time resolution which is critical for investigating turbulent flow fields such as those in many plasma and combustion environments.

The schematic of a typical RHI breadboard used for many of our demonstrations is shown in Figure 1. For the simultaneous recording of a two-wavelength resonance hologram, the timing of the on-resonance λ_1 and off-resonance λ_2 pulses are overlapped. The temporal overlap is accomplished through optical delay lines. The temporally overlapped laser beams are collinearly combined at beam combining element ***BC***. The combined beam is split by beamsplitter ***BS*** forming an object and reference beam which is composed of both on-resonance and off-resonance wavelengths. The object beam is directed through the probed medium. The reference beam

traverses a distance equal to that of the object beam, but bypasses the probe volume. The object beam and the reference beam are directed to a holographic film plate where they interfere to produce the resonance hologram. Pulse energy requirements to expose a 50 mm diameter hologram (Kodak 120, or Agfa Gavaert 8E56) are on the order of 1 mJ in an 8 ns pulse.[1,2], These requirements are easily met with commercial pulsed dye lasers.

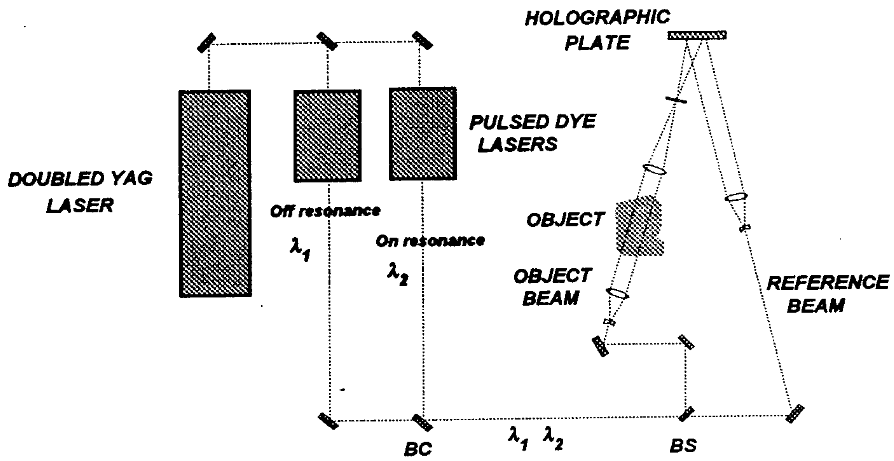

Figure 1. . Schematic diagram of RHI instrument.

RESULTS

The RHI experimental breadboard was configured for operation in three wavelength regions: visible, UV, and near IR.. In the course of investigating these spectral regions, opportunities for technological advances were recognized and preliminary experiments were performed in the areas of combusting sprays, stimulated Brillouin scattering, and data reduction methods. These results are outlined below.

Visible

We began our examination of RHI applications in the visible spectral region. For this study we probed three different environments. In increasing order of complexity, we made RHI measurements of I_2 in a heated sample cell, Na in a seeded methane - air flame, and C_2 in an arcjet torch reactor.

I_2 Static Test Cell

In an effort to produce quantitative RHI results, an experiment was set-up utilizing a wedge-shaped static test cell. Iodine was placed in the test cell which was then inserted into a tube furnace to provide precise control of the vapor pressure, and thus the amount of I_2 in the gas phase. For constant volume conditions, the vapor pressure is directly proportional to the temperature of the cell and therefore, a precise amount of I_2 may be introduced into the object path of the RHI instrument. The wedge shape of the test cell provided a known pathlength variation over the field of view. A diagram of the I_2 cell and tube furnace is shown in Figure 2.

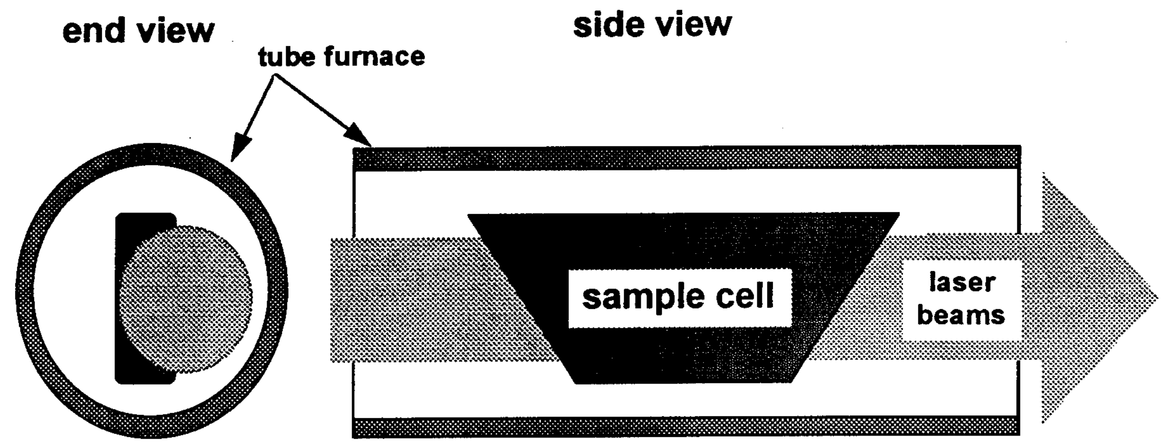

Figure 2. Schematic diagram of static I_2 test cell and tube furnace.

In Figure 3 we show the qualitative results for the I_2 experiment. The results are not quantitative due to poor reproducibility. The difficulty resulted from multiple longitudinal mode effects caused by dye laser cavity instability. The multi-mode behavior adversely affected laser linewidth, and hence our ability to consistently generate data sets with consistent fringe shifts. Subsequent experiments in the UV and near IR spectral regions used a more stable dye laser, and therefore, the dye laser cavity instabilities were less of a problem.

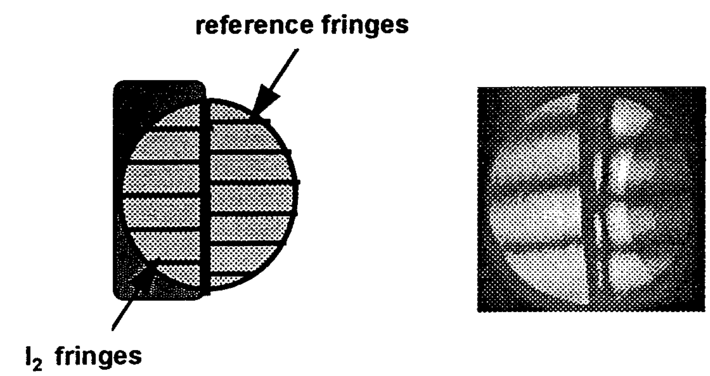

Figure 3. RHI interferogram of I_2 in a static test cell.

Na-seeded Methane/Air Flame

The first successful combustion RHI experiments were carried out in the flame of a CH_4-air diffusion burner. This experiment was designed to unequivocally demonstrate the RHI effect. High quality, Na RHI interferograms were recorded in this flame environment. Demonstrations of the RHI technique in a Na-seeded and unseeded CH_4-air flame are presented below. Figure 4 illustrates the geometry of the flame, imaged area, and flowfield seeding method.

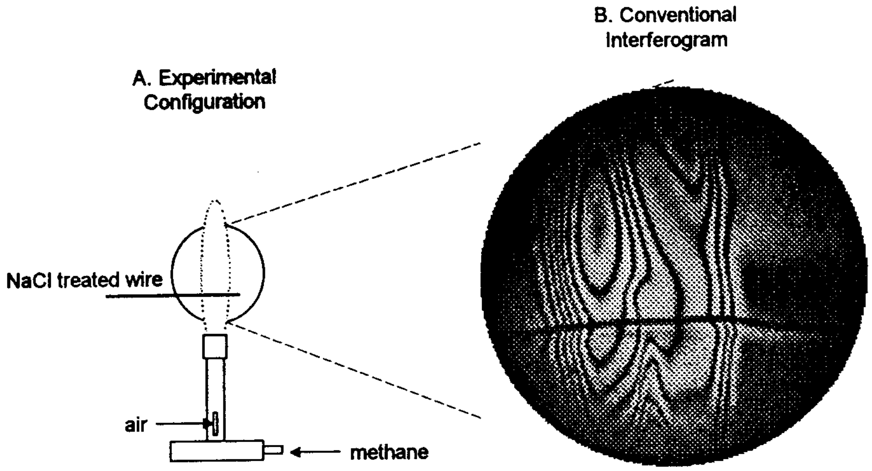

Figure 4. (a) Experimental configuration. (b) Conventional holographic interferogram.

In the configuration shown in Figure 4a, a CH_4-air burner was seeded with Na by introduction of a pre-treated wire approximately one third of the way into the flame. This configuration generates both seeded and unseeded flame regions which are recorded in a single image. The wire had been pre-treated by immersion in saltwater. Na^+ ions from the treated wire entered the hot combustion gases through vaporization and ablation and were rapidly neutralized by free electrons in the flame, resulting in the formation of Na atoms. The strongest Na atomic line D_2 (j=3/2) was used for probing at approximately 589.015 nm. Intense yellow-orange emission was clearly visible by eye in the Na-seeded portion of the flame above the screen. In the unseeded portion of the flame below the wire, the only emission visible by eye was blue.

Figure 4b shows a reconstruction of a conventional holographic interferogram. In this case, a single laser beam (tuned to 588.972 nm, a wavelength which is well off the D-line resonance) was used to double expose the holographic plate at two instances in time. The first exposure was taken with the flame on. The second exposure was taken with the flame off. The resultant fringe pattern shows the combined effects of variations in bulk density, temperature, and pressure associated with all the species present in the flame.

In contrast to this conventional hologram, Figure 5a shows a single exposure holographic interferogram with the flame on, taken with two simultaneous laser pulses of the same wavelength (both tuned off resonance to 588.972 nm). This demonstrates that phase variations due to temporal fluctuations and bulk density gradients are completely subtracted out in the holographic reconstruction process.

A. B.

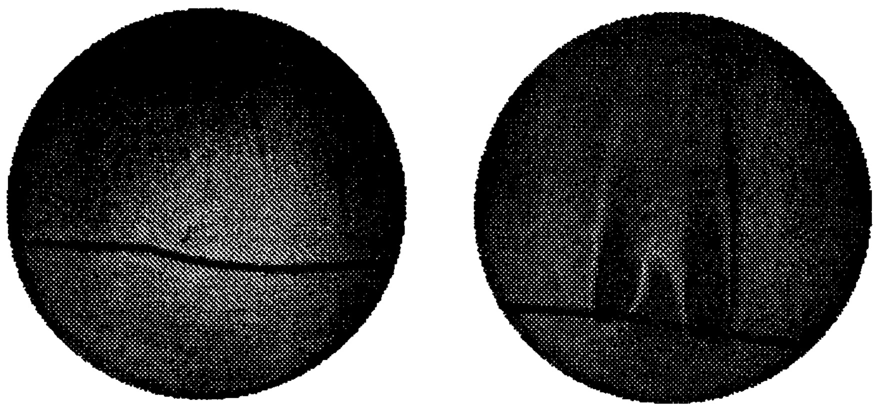

Figure 5. (a) Two-wavelength off-resonance interferogram of flame demonstrating total background subtraction inherent to holographic reconstruction. (b) RHI interferogram of Na seeded into a CH_4-air diffusion burner flame.

Figure 5b is a RHI interferogram of the Na target species in the flame. The two lasers were tuned symmetrically to either side of the Na D_2 line (589.009 nm and 589.021 nm). The fringes are due solely to the Na variation in the flame. In the lower part of the flame below the wire and upstream of the Na seeding, there are no fringes present due to the absence of Na and the complete subtraction of the background refractive index variations. Because of the high concentration of Na atoms in the seeded portion of the hot inner cone of the flame, regions of total absorption are evident. These results provide information regarding both the minimum detectable concentration for a given path length and molecular species, and the range of concentration values that can be detected in a single hologram.

C_2 in Arcjet Plume

Attempts at obtaining RHI interferograms of diatomic carbon in an arcjet torch were unsuccessful. An arcjet torch running on a 10 to 20 torr mixture of Ar, H_2, and CH_4, was probed by the RHI instrument. The RHI lasers were tuned in the vicinity of the bandhead near 516 nm. Our goal was to resolve the spatial distribution of C_2 in the high temperature plasma. Initial estimates of the C_2 number density in the plasma indicated that RHI would have the necessary sensitivity for its detection; however, since no RHI fringe shifts were detected, we were not able to verify these predictions. We concluded that the low pressure arcjet environment does not have appreciable C_2 number densities for detection by single-pass RHI methods.

Ultraviolet (UV)

Since many important combustion species exhibit absorptions in the UV spectral region, we reconfigured the RHI breadboard for examinations at these shorter wavelengths. We chose the hydroxyl radical as the target species for the UV investigations for several reasons. Hydroxyl is an important combustion intermediate that is often used for thermometry, it is useful for marking the reaction zone, and its spectroscopy is well known. The environment we chose was the methane - air diffusion flame produced from a laboratory slot burner.

OH in Methane/Air Flame

RHI interferograms were recorded for species naturally occurring in flames. Figure 6 shows a reconstructed interferogram of OH radicals in a CH_4-air diffusion flame. For this work, the RHI lasers were tuned in the vicinity of the bandhead near 308 nm.. The single dark fringe visible in Figure 6 uniquely corresponds to the location of OH in the post flame gases and is an indicator of OH number densities on the order of 10^{15} cm^{-3}. This result agrees well with our theoretical estimates and observations reported by others[3].

RHI of OH $X^2\Sigma^+ - A^2\Pi$ (0,0)

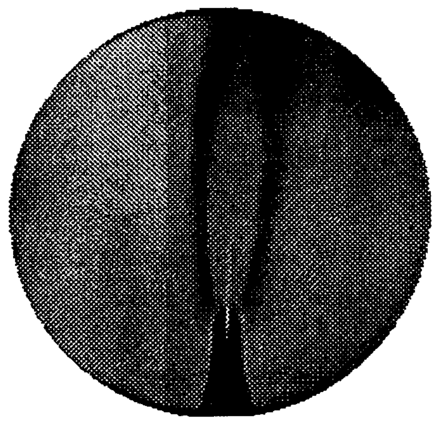

Figure 6. RHI interferogram of OH in methane-air diffusion flame.

Near Infrared (near IR)

One of the primary attractions for examining RHI applications in the near IR spectral region is the potential use of inexpensive, compact laser diodes. In order to examine RHI applications in the near IR spectral region, we substantially modified the RHI holocamera to accommodate photorefractive recording media. Several important benefits are gained by using photorefractives for the RHI holographic recording medium. Photorefractives allow real-time writing, reading, and erasing capability. The ability to collect and process the data in real-time facilitates experimental alignment. Photorefractives are also less susceptible to fogging from incoherent background radiation.

For our RHI examination in the near IR, we doped fuel (methanol) with potassium (KCl). The potassium was used to mark the flame front boundary of the combusting fuel droplet.

Real-time RHI of K using Photorefractives

Demonstration RHI experiments using novel photorefractive materials were performed. New photorefractive crystals make it possible to not only generate RHI interferograms in the near IR spectral region, but to generate them at video framing rates (30 Hz). As one example of this work, Figure 7 shows a single frame of an RHI interferogram of a KCl-seeded methanol droplet undergoing combustion. The droplet was suspended on the end of a fine wire and ignited. The RHI lasers were tuned near 766 nm. When potassium ions are released from the methanol, they become neutralized by free electrons in the flame front reaction zone. The neutral potassium atoms are a clear marker for the flame front. The fringes due to potassium in the flame were traced manually and digitized. The integrated number density is calculated and displayed to the right of the image.

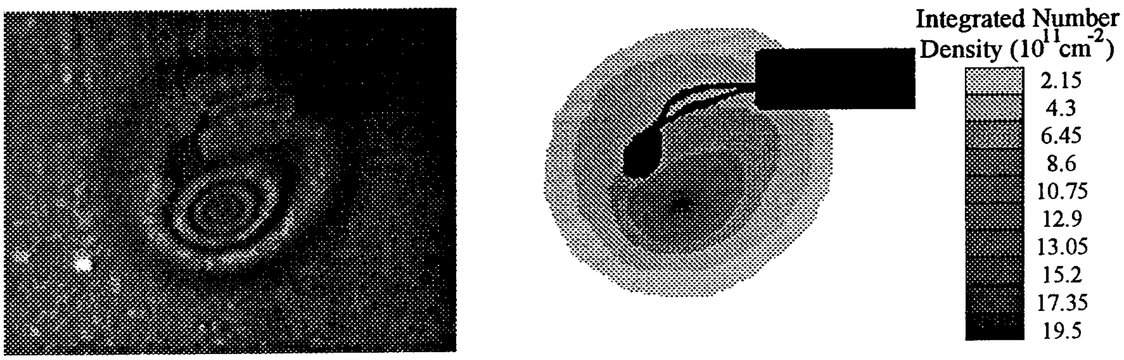

Figure 7. Real-time RHI of potassium seeded combusting 1 mm methanol drop.

TECHNICAL OPPORTUNITIES IDENTIFIED

In the course of our RHI development work, several opportunities were recognized. Below we highlight three opportunities discovered: combusting sprays, stimulated Brillouin scattering, and quantitative data reduction.

Combusting Sprays

MetroLaser performed preliminary investigations relative to the use of RHI in droplet combustion.[4] By utilizing photo-refractive recording materials to write and read holograms in real time, we have produced an RHI video movie of fuel combustion in mono-disperse droplet streams and sprays. Figure 8 shows three frames from the video. The RHI fringes marking the flame front boundaries indicate that the droplet spacing is increasing from the left frame to the right frame.

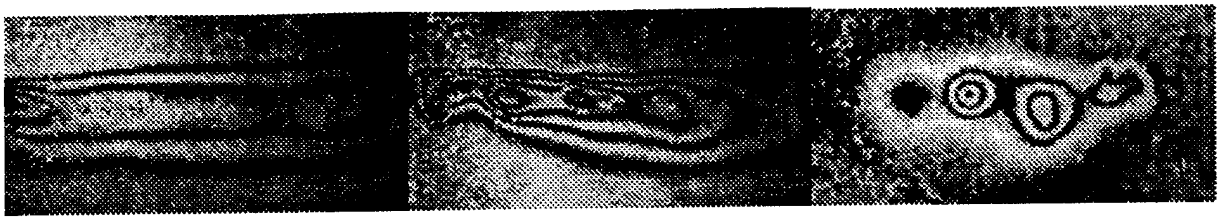

Figure 8. RHI interferograms of combusting fuel droplet streams. Droplet size is approximately 100 microns.

Stimulated Brillouin Scattering

In order to reduce the cost and complexity of an RHI laser system, techniques were investigated for performing the experiments with a single dye laser. By frequency shifting a portion of a tunable dye laser beam in a stimulated Brillouin scattering (SBS) medium, a second phase conjugate beam is generated. By judiciously choosing the SBS medium, the amount of frequency shift can be made to coincide with that needed to symmetrically straddle an absorption line. For example, frequency shifting a 308 nm laser beam in liquid hexane generates a phase conjugate beam that is shifted by approximately 0.25 cm^{-1}. This amount of shift fortuitously corresponds with the width of the Doppler broadened OH transition at flame temperatures, and is thus ideally suited for single dye laser RHI applications. As a demonstration of this, we adapted the UV RHI breadboard to include an SBS cell for frequency shifting one of the RHI dye lasers. Figure 9 shows an RHI image of OH in methane-air diffusion flame that was recorded using a single dye laser and an SBS cell. The slight fringe shift observed in the vicinity of the reaction zone is consistent with OH number densities on the order of 10^{14} cm^{-3}.

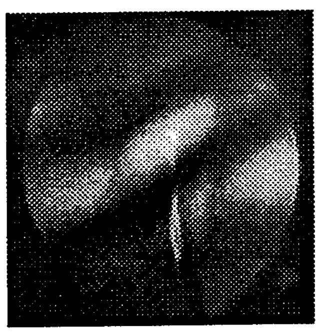

Figure 9. Image of OH in CH_4-air diffusion flame recorded with RHI-SBS configuration.

Quantitative Data Reduction

Since the observed RHI fringe shifts are typically a very small fraction of one complete fringe, a very sensitive method for recording the fringes, and a very accurate method for fringe tracking and data reduction is needed for quantitative analysis. To these ends, we examined the technique of phase shifting interferometry (PSI) only to discover physical limitations of the technique preventing its use with RHI. Consequently a new technique, employing a correcting holographic optical element (CHOE), was developed to overcome these physical limitations thus allowing the method of PSI to be used with RHI. The following sections describe PSI, the problems

encountered when using PSI with RHI, and the CHOE concept developed to eliminate these problems.

Phase Shifting Interferometry

PSI is a versatile technique for reducing an interferogram into a raw phase map with high spatial resolution. PSI is a well developed technique that is widely used in the field of optical testing[5]. The sub-fringe sensitivity, which is capable with phase shift interferometry, is extremely desirable for RHI since in many applications the resonant species under investigation will produce only a small fraction of a phase shift.

In PSI, a small phase shift is introduced between the reference and object beam of an interferometer. The resulting interferogram is then digitized for image processing by computer. At least three (or four) incrementally phase-shifted interferograms are required for each test object being studied. The encoded phase map of the test object is contained in the set of digitized interferograms. Several computer algorithms have been developed for extracting the phase map from the set of interferograms.

A RHI system adapted for PSI requires two reference beams. During reconstruction, a known phase shift is added to one of the reference beams. This process is repeated to generate three different interferograms from each hologram. The data reduction codes for PSI read the intensity distributions from each interferogram, compute a phase map of the optical wavefronts, and convert the phase map into a concentration distribution. With PSI, it is possible to map the phase of the interferogram with an accuracy of hundredths of a fringe.

Problems with using PSI with RHI

In RHI, the recordings are often made with UV lasers; however, reconstructions must be done with visible lasers because of the UV absorbing holographic medium.[6] Reconstructing the multiple images required in PSI can also be difficult with the pulsed recording laser because pulse to pulse variations can introduce image variations which result in phase errors when processed by PSI. We found that the combinations of large angle change and aberrations caused by the wavelength difference between recording and reconstruction, combined with high optical quality and alignment tolerances, made two reference wave, phase shift interferometry impractical with conventional methods. The resulting interferograms contained many complex (aberration) interference fringes, rendering them unusable. A method was needed to store and optically subtract such effects during the reconstruction process. The method we developed employs holography to compensate optically for the problems of reference wave lens aberrations and misalignment and chromatic aberrations. This method utilizes a device called a Correcting Holographic Optical Element (CHOE) which corrects for aberrations resulting from necessary differences between the optical systems used for construction and reconstruction.

Correcting Holographic Optical Element (CHOE)

A CHOE is simply a hologram which is used to correct for aberrations that result when a two-reference wave hologram is reconstructed using a different wavelength and optical set-up than that used during the making of the hologram. The concept of the CHOE technique is straightforward. First, the data hologram is recorded with the object present using two reference beams. In the second step, the CHOE is recorded with the object removed and a slight angular change made to the object beam. The data and CHOE holograms are then sandwiched together utilizing a double plate holder. When the CHOE is reconstructed with a replica of the object beam, the stored reference beams emerge at an angle that is wavelength corrected for reconstructing the data hologram without introducing chromatic aberrations. Since the reconstructed reference beams contain identical aberrations introduced by the reconstruction optics, the aberrations cancel. Since both the data and the CHOE holograms contain identical aberrations introduced during hologram recording, these aberrations also cancel. The resulting interferometric image emerging from the data hologram is completely aberration free. Phase shifting of the interferometric image, which is required by PSI, is simply accomplished by rotating the sandwiched plates during reconstruction.

An experiment performed to test the CHOE concept demonstrated that the method appears to eliminate all of the problems encountered when using different optical systems and different lasers between hologram construction and reconstruction. This method enables the technique of PSI to be used with RHI at any wavelength and with virtually any optical system for interferogram data reduction.

CONCLUSIONS

During this three-year program, MetroLaser pioneered the development of RHI; a holographic technique capable of recording the concentration of a target species in complex flame environments. Some of the attributes that make RHI attractive to the combustion community are:

- Insensitive to quenching,
- background free signals, and
- simultaneous imaging of gaseous species and solid or liquid object.

The program targeted the measurement of species with resonant transitions in the visible, UV, and near IR. This broad operating spectrum required the development of auxiliary technologies such as real-time holographic recording and reconstruction using nonlinear optical materials. For the first time, holographic movies were made that followed the combustion of a target species in a flame. Nonlinear optical materials opened another door of opportunity: IR holography. Until recently, holograms were limited to wavelengths below 700 nm due to limitations of the recording media. Photorefractive semiconductors have broken this barrier resulting in high quality holographic recordings out to 2 μm. This expanded spectral operating range coincides with commercial diode laser wavelengths. This move into the near IR region establishes a pathway toward commercialization of this technology.

RHI can simultaneously record the concentration of a gaseous species and the image of a liquid or solid object. For example, burning fuel droplets can be recorded showing the droplet and a selected gaseous species. The image of the droplet may be used to assess fuel evaporation while the gaseous species concentration may be an indication of flame front position or mixing scales.

The most pressing challenge which remains is in the development of RHI to convert the holographic images into quantitative species concentration profiles. Although RHI recordings can be easily inspected by eye to observe qualitative features of the combustion process, computerized data reduction and sub-fringe resolution are considerably more difficult to implement. To date, only estimates of the species concentration have been made based upon manual fringe interpretation.

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